



(19)

Europäisches Patentamt

European Patent Office

Office européen des brevets



(11)

EP 0 747 521 A2

(12)

EUROPEAN PATENT APPLICATION

(43) Date of publication:  
11.12.1996 Bulletin 1996/50

(51) Int Cl.<sup>6</sup>: D04H 1/54

(21) Application number: 96304104.1

(22) Date of filing: 05.06.1996

(84) Designated Contracting States:  
DE GB IT

(72) Inventor: Terakawa, Taiju  
Yasu-gun, Shiga-ken (JP)

(30) Priority: 06.06.1995 JP 164749/95

(74) Representative: W.P. THOMPSON & CO.  
Eastcheap House  
Central Approach  
Letchworth, Hertfordshire SG6 3DS (GB)

(54) Continuous fiber nonwoven and method for producing the same

(57) A continuous fiber nonwoven comprising composite continuous fibers having spiral crimps obtained by compositely spinning two thermoplastic resins having a difference in the melting points of 15°C or more is

provided. It is characterised in that the contact points of the fibers are adhered to one another by fusing of the thermoplastic resin having the lower melting point and located on the outside of the spiral crimps.

**Description**

The present invention relates to a continuous fiber nonwoven produced by heat fusion and having excellent bulkiness and high tensile strength. More particularly, the invention provides a continuous fiber nonwoven usable for sanitary materials, engineering materials, agricultural materials, packing materials and the like.

In methods for producing nonwovens utilizing characteristics of heat fusion, there is known a heat treating method of carding webs comprising staple fibers and also a heat treating method of continuous fiber webs. Although the latter method has the advantage that the production process is simple, the resulting nonwoven has the disadvantages of low flexibility and low bulkiness.

Conventional continuous fiber nonwovens, which are produced by a method of heat fusion and are usable for sanitary materials, engineering materials and the like, are mainly made of fibers of one component. Since such fibers do not develop crimps, they have low bulkiness.

Known methods for developing the steric crimps of a spiral form (abbreviated as spiral crimps hereinafter) in the fibers of one component, include a method for developing the spiral crimps based on the difference of heat shrinkage inside the fiber by pulling out the spun fiber while partial quenching is applied to the fiber (Japanese Patent publication No. 45-1649), and a method for developing the crimps based on the difference of the degree of crystallization by blending a nucleating agent into a certain part of the fiber cross-section (Japanese Patent Application Laid-open No. 5-209354). In the former method, however, the crimps are loosened through the heat treatment process for processing the fiber into a nonwoven and the bulkiness becomes insufficient. In both methods, since the fiber is constituted from one component, a hot pressing method is only used as the heat treatment process for processing the fiber to the nonwoven, so that the spiral crimps of the fiber are pressed, resulting in undesirable bulkiness.

It is known that spiral crimps can be developed in the fiber by compositely spinning several thermoplastic resins into a parallel or eccentric sheath core type arrangement (Japanese Patent Applications Laid-open Nos. 48-1471 and 63-282350). In the nonwovens using these composite fibers, however, although it is recognized that the bulkiness is improved, the tensile strength is the same as (or less than) that of conventional nonwovens made of one component fibers, so that more improvement has been sought.

The present invention provides a continuous fiber nonwoven having excellent bulkiness and high tensile strength in view of the above conditions of the continuous fiber nonwovens produced by heat fusion methods.

The present invention seeks to solve the aforesaid problems by aiming at the relationship between the spiral crimps developed in the composite fibers and the arrangement of components on the fiber cross-section. These aims are attained by using composite fibers comprising several thermoplastic resins arranged in a parallel or eccentric sheath core type, in which the thermoplastic resin having a lower melting point is located on the outside of the spiral crimps developed by stretching the fibers.

In accordance with a first aspect of the present invention, there is provided a continuous fiber nonwoven comprising composite continuous fibers having spiral crimps obtained by compositely spinning two thermoplastic resins having a difference in melting point of 15°C or more, characterised in that the contact points of the fibers are adhered to one another by fusing of the thermoplastic resin having the lower melting point and located on the outside of the spiral crimps.

Also in accordance with the present invention there is provided a method for producing a continuous fiber nonwoven comprising: preparing a first thermoplastic resin and a second thermoplastic resin having a melting point at least 15°C less than that of the first thermoplastic resin and an elastic shrinkage 1% less than that of the first thermoplastic resin; compositely spinning these resins in a composite ratio of 60/40 - 40/60 into a parallel type or an eccentric sheath core type, in which the second thermoplastic resin is a sheath and the first thermoplastic resin is a core eccentric to the sheath; stretching the resulting yarn over 1.2 times as long as the unstretched yarn at a temperature lower than the melting point of the second thermoplastic resin; and heat treating the yarn at a temperature higher than the melting point of the second thermoplastic resin and lower than the softening point of the first thermoplastic resin to adhere one to the other at the contact points of the fibers.

The thermoplastic resins used as raw materials of composite continuous fibers can include, for example, polyolefins such as polypropylene, polyethylene, ethylenepropylene copolymer, propylene-butene-1 copolymer, ethylene-propylene-butene-1 copolymer, ethylene-vinyl acetate copolymer, and poly-4-methylpentene-1, polyolefins modified with unsaturated carboxylic acids or their anhydride, polyesters such as polyethylene terephthalate, polyethylene terephthalate-isophthalate copolymer and polybutylene terephthalate, polyamides such as nylon 6, nylon 66 and nylon 12, thermoplastic polyurethane and the like.

In the present invention, a combination of two kinds of thermoplastic resins having a difference in melting point of 15°C or more is selectively used. In this case, it is necessary to use spinning conditions so that the elastic shrinkage of the thermoplastic resin having a higher melting point becomes 1% or more higher than that of the thermoplastic resin having a lower melting point.

With the present invention, nonwovens are obtained by heat treating the composite continuous fibers and adhering the contact points of fibers by fusing only thermoplastic resin having a lower melting point. If the difference of the

melting points of two thermoplastic resins, which are raw materials of composite fibers, is less than 15°C, this is undesirable because the temperature range usable in the heat treatment becomes narrow.

The term "elastic shrinkage" means a shrinkage that unstretched yarn of one component is stretched to the same draw ratio (K) as drawing conditions of the composite fibers and at once the load is removed, and the following equation is provided.

$$\text{Elastic shrinkage } S(\%) = 100 \times (KA-B)/(KA-A)$$

A: length of unstretched yarn

B: length of yarn at removal of load after stretching the yarn.

When it is impossible to spin one component fiber of thermoplastic resin (a), or it is impossible to stretch it to the length of 1.5 times, elastic shrinkage (S1) of the unstretched yarn composed of a single component of thermoplastic resin (b) having excellent stretch properties, and elastic shrinkage (Sc) of the unstretched yarn composite fibers composed of thermoplastic resin (a) and thermoplastic resin (b), are measured, and the elastic shrinkage (S2) of the unstretched yarn of thermoplastic resin (a) is calculated by the following equation:

$$S2 = 2Sc - S1$$

When the difference of the elastic shrinkages of two thermoplastic resins is less than 1%, distinct crimps are not observed after stretching the composite fibers, and one cannot obtain sufficiently bulky nonwovens. In the case of two thermoplastic resins, if the elastic shrinkage of the thermoplastic having a higher melting point is less than that of the thermoplastic resin having a lower melting point, it is impossible to locate the thermoplastic resin having a lower melting point on the outside of the spiral crimps which appear after the composite fibers are stretched.

In the composite continuous fibers used in the present invention, two thermoplastic resins selected in accordance with the above standards are preferably compositely spun into a parallel type or an eccentric sheath core type in the range of a composite ratio of 60/40 - 40/60. Since the crimps of the composite fibers are based on the difference between the elastic shrinkages of both components, clear crimps do not appear when one component is present at less than 40%, so that sufficiently bulky nonwovens are not obtainable. In case of the eccentric sheath core type, thermoplastic having the lower melting point is used at the sheath side of the composite fibers.

Crystalline polypropylene/polyethylene can be exemplified as a desirable combination of two thermoplastic resins, and crystalline polypropylene having a wide molecular weight distribution can desirably be used as a thermoplastic resin having a high melting point, because it shows a relatively high elastic shrinkage.

After the unstretched yarn obtained by the composite spinning is stretched, and immediately the stress is removed, the spiral crimps develop in the composite fibers. The curvature radius of the spiral is based not only on physical properties of the differences between the elastic shrinkages of the raw material resins, the Young's modulus, the fineness and the like, but also on the stretching temperature and the draw ratio. The stretching conditions are selected in accordance with the degree of bulkiness of desired nonwovens (commonly 1.2 - 4 times the length of unstretched yarn, between room temperature and a temperature lower than the melting point of the second thermoplastic resin).

In such obtained composite continuous fibers, the thermoplastic resin having a lower melting point is located on the outside of the spiral crimps.

To obtain the web of the composite continuous fibers having spiral crimps and used in the present invention, two thermoplastic resins selected in accordance with the said standards are compositely spun at the fixed composite ratio, and the unstretched yarn stored on bobbins or in canes is stretched under the fixed stretching conditions and is immediately accumulated on a conveyor. It is also possible to use a spunbond method in which the spun composite fibers are pulled by a stretch machine equipping a feed roll and a draw roll via a quenching device, and then accumulated on a conveyor net in which the fibers are sucked with an air sucker and the fibers are opened.

The continuous fiber nonwoven of the present invention can be obtained by heat treatment of the above composite continuous fiber webs having spiral crimps at a temperature higher than the melting point of the thermoplastic resin having the lower melting point and lower than the softening point of the thermoplastic resin having a higher melting point. In the heat treatment, a hot pressing device such as an embossing roll, or a suction dryer with internal air circulation, or a heater such as an infrared heating oven, may be used.

Although the contact points of the fibers are adhered by heat treatment to fuse the thermoplastic resin having a lower melting point, because the thermoplastic resin having a lower melting point is located on the outside of the spiral crimps in the composite continuous fibers used in the present invention, the fibers contact one another by the thermoplastic resin having a lower melting point, the fibers are adhered to one another by fusion of the same kinds of ther-

moplastic resins, and nonwovens having a high tensile strength are obtained.

When a hot pressing device is used in the heat treatment, the temperature of the heat treatment may be a temperature near the softening point of the thermoplastic resin having a lower melting point, which is located on the outside of the spiral crimps, so that the thermoplastic resin having a higher melting point does not soften or change the shape by heat, and bulky and soft nonwovens can be obtained.

To obtain nonwovens having a sufficient strength by using the composite fibers in which thermoplastic resin having a lower melting point is located on the inside of the spiral crimps, it is necessary to treat the fibers at higher temperature to soften the thermoplastic resin having a higher melting point, so that the touch of the nonwoven becomes hard.

Since the suction dryer with internal air circulation can provide a sufficient heat capacity without pressing its continuous fiber web, it is preferably used for producing bulky nonwovens at a high speed. In this case, since the thermoplastic resin having a lower melting point is located on the outside of the spiral crimps, the composite fibers contact one another with the thermoplastic resin having a lower melting point to fix the fibers by fusing the same kind of thermoplastic resins, and nonwovens having a high tensile strength are obtained.

When the fibers are heated at a temperature to fuse the thermoplastic resin having a lower melting point, the thermoplastic resin having a higher melting point slightly shrinks to reduce the strain produced by stretching the fibers, while the thermoplastic resin having a lower melting point greatly shrinks and fuses, and as a result, the spiral crimps reversely turn so as to arrange the thermoplastic resin having a high melting point outside the spiral crimps of the composite fibers. By such fibers, the numbers of contact and adhered points among the fibers are increased, thereby to obtain nonwovens having a high strength. Further, since the fibers pull one another between the adhered points, the bulkiness is little decreased.

When the composite fibers, in which the thermoplastic resin having a lower melting point is arranged on the inside of the spiral crimps, are heat treated with a suction dryer, the spiral crimps of the composite fibers become smaller by the shrinkage and the fusing of the thermoplastic resin having a lower melting point, the bulkiness of the nonwoven is lost, and the strength of the nonwoven decreases with the decrease in number of the adhered points among the thermoplastic resins having a lower melting point.

Since the continuous fiber nonwoven of the present invention is obtained by using the composite continuous fibers as raw fiber materials in which the thermoplastic resin having a lower melting point is located on the outside of the spiral crimps, it has the same or a higher degree of tensile strength in comparison with conventional nonwovens of continuous fibers, and it has high bulkiness which is not observed in the conventional nonwovens. Accordingly, it is possible to use the nonwovens of the present invention as sanitary materials for surface materials of diapers and the like, geotextile materials, packaging materials, etc.

The present invention is illustrated more specifically by the following Examples and comparative Examples. The physical values in these Examples are determined by the following methods.

#### 35 Elastic shrinkage:

An unstretched yarn of one component fibers and an unstretched yarn of composite fibers are stretched at a grip distance of 10 cm and a stretching rate of 10 cm/min to the same magnification (K) in Examples and comparative Examples, and these yarns are immediately returned to the beginning grip distance, the fiber length (c) of a zero point of the stretching load is measured and then the elastic shrinkage (S) is calculated by the following equation:

$$\text{Elastic shrinkage } S (\%) = 100 \times (10K - c)/(10K - 10) \quad S2 = 2Sc - S1$$

#### 45 Arrangement of components of spiral crimps:

A specimen having one cycle length of the spiral crimps is cut off from the composite fibers, it is put between two pieces of cover glass to form a circle, and observing the melting behavior of the thermoplastic resin having a lower melting point by using an optical microscope equipping a hot stage, the arrangement of components is identified.

#### 50 Number of crimps:

A fiber having ten spiral crimps is cut off and the straight length L (cm) is measured and the number of crimps is calculated using the following equation:

$$\text{Number of crimps (crimps/inch)} = 10 \times 2.54/L$$

Specific volume of nonwoven:

5 Four test pieces having 10 cm length and 10 cm width are piled, a plate having the same length and width and 20 g weight is put on the test pieces, the thickness D (cm) of the four test pieces is measured, the total weight W1 (g) of the four test pieces is previously measured, and the specific volume of nonwoven is calculated by the following equation:

$$\text{Specific volume of nonwoven (cm}^3/\text{g}) = 100 \times \text{D/W1}$$

10 Tensile strength of nonwoven:

15 Test pieces having 20 cm length and 5 cm width (weight is W2) are cut off from nonwoven in a machine direction of nonwoven production (MD) and its cross direction (CD), maximum load power P (g) is measured at a grip distance of 10 cm and a stretching rate of 10 cm/min, and the tensile strength is calculated using the following equation after gr/m<sup>2</sup> is corrected:

$$\text{Tensile strength (g/(cm} \times \text{g/m}^2)) = P/500W2$$

20 Geometric mean strength = (MD strength x CD strength)<sup>1/2</sup>

Examples 1 to 5 and comparative Examples 1 to 4:

25 Table 1 shows the production conditions of raw continuous fibers and properties of the continuous fibers used for nonwovens of the Examples and the comparative Examples.

30

35

40

45

50

55

5  
10  
15  
20  
25  
30  
35  
40  
45  
50  
55

(Table 1)

	Spinning and stretching conditions						Properties of fibers					
	Fiber components	Temp. of extruder	Spinneret temp.	Elastic shrinkage %	Difference of elastic shrinkage %	Fineness d	Stretch temp. °C	Stretch ratio	Arrangement outside/inside	Number of crimps	Yarn strength g/d	Yarn elongation %
Example 1	HDPE parallel PP1	240	280	25.2	2.6	2.0	room temp.	2.0	HDPE PP1	6.5	2.43	169
	HDPE parallel PP2	290	280	27.8	7.0	2.0	room temp.	2.0	HDPE PP2	12.0	2.25	180
Example 3	HDPE eccentric sheath core PP2	240	280	25.2	7.0	2.0	room temp.	2.0	HDPE PP2	9.5	2.12	195
	HDPE eccentric sheath core PP2	310	310	32.2	36.7	2.0	room temp.	1.7	HDPE PP2	11.0	1.88	224
Comparative example 1	PP1 only one	290	260	27.8	...	2.0	room temp.	2.0	...	0	2.71	136
	HDPE Parallel PP2	240	280	25.2	0.8	2.0	...	...	PP1 HDPE	7.8	1.38	275
Comparative example 2	HDPE Parallel PP2	290	280	26.0	...	...	...	...	Developing poor crimps	2.0	1.98	206
Comparative example 3	HDPE Parallel PP2	240	280	25.2	0.8	2.0	room temp.	2.0	...	...	...	...

Note: PP1 = Crystalline polypropylene, MFR=10, Q=3.5, m.p.=164°C, s.p.=144°C

PP3 = Crystalline polypropylene, MFR=25, Q=5.0, m.p.=164°C, s.p.=143°C

HDPE = High-density polyethylene, MFR=40, m.p.=129°C, s.p.=100°C

Fibers of Examples 1 to 3, which were obtained by combining crystalline polypropylene and high-density polyethylene, compositely spinning them, and stretching the yarn, have developed desirable spiral crimps by arranging 5 high-density polyethylene outside the spiral crimps. In Example 2, composite fiber having many crimps is obtained by the same conditions of spinning and stretching as in Example 1. It is considered that the fact is caused by using crystalline polypropylene having wide molecular 10 weight distribution (high Q value).

Although the composite fiber, which was obtained in Example 3 by using the same raw materials, spinning temperature and stretch conditions as in Example 2, develops desirable spiral crimps arranging high-density polyethylene, the number of crimps is less by changing the composite type to an eccentric sheath core type. However, by changing the stretch conditions, it was possible to obtain the composite fiber of the eccentric sheath core type having many crimps (Example 4).

The fiber comprising one component of crystalline polypropylene (comparative Example 1) does not develop the spiral crimps even though the fiber was stretched as in Example 1.

In comparative Example 2, in which the fiber was extruded by using the same conditions as in Example 1 and directly spun with an air-sucker instead of machine stretching, the fiber developed spiral crimps, on the inside of which high-density polyethylene, the component having a low melting point, was arranged.

In comparative Example 3, in which the composite fiber was obtained by spinning and stretching the yarn by the same process as in Example 1 except that the extrusion temperature of crystalline polypropylene was increased, the difference of elastic shrinkages became smaller and very poor spiral crimps were developed.

The webs of various continuous fibers were processed by heat treatment with a heat oven with internal air circulation or a heat embossing roll to obtain nonwovens. The process conditions and the physical properties of the nonwovens are shown in Table 2.

25

30

35

40

45

50

55

(Table 2)

	Processing conditions			Physical properties of nonwoven		
	Air circulation oven temperature	Emboss. temp.	Basis weight g/m <sup>2</sup>	Thickness mm	Specific volume cm <sup>3</sup> /g	Geometric mean strength (*)
Example 1	135°C 1.7 sec.	-- --	30	1.19	39.8	26.3
Example 2-1	135°C 1.7 sec.	-- --	30	1.46	48.8	26.0
Example 2-2	--	125°C 15%	30	0.70	23.3	28.0
Example 3	135°C 1.7 sec.	-- --	30	1.37	45.6	27.3
Example 4	135°C 1.7 sec.	-- --	30	1.40	46.5	24.8
Comparative example 1	-- --	145°C 15%	31	0.26	8.5	30.0
Comparative example 2-1	135°C 1.7 sec.	-- --	31	0.94	30.3	12.2
Comparative example 2-2	-- --	125°C 15%	30	0.51	17.0	15.5
Comparative example 3	135°C 1.7 sec.	-- --	30	0.46	15.2	25.4

(\*) : g/(cm · g/m<sup>2</sup>)

The nonwoven comprising one component fiber of crystalline polypropylene obtained in comparative Example 1 is poorer in bulkiness and strength than those of the other Examples.

The nonwoven prepared in comparative Example 2-1 by using the same raw materials and process conditions as in Example 1 is poor in bulkiness (thickness and specific volume) and strength in comparison with the nonwoven in Example 1. It is considered that the fact is caused by arranging crystalline polypropylene having elastic shrinkage outside of the spiral crimps, and by arranging high-density polyethylene having adhesion properties on the inside of

the spiral crimps.

The nonwoven prepared by a heat embossing roll in Example 2-2 is poor in bulkiness, but it is good in strength in comparison with the nonwoven obtained in Example 2-1. The nonwoven of Example 2-2 is good in both bulkiness and strength in comparison with the nonwoven prepared by the heat embossing roll in comparative Example 2-2.

5 Although the raw materials are different from those in Example 1, the nonwovens of Examples 3 and 4, in which the difference of the elastic shrinkage and the constitution of the spiral crimps satisfy the requirements of the present invention, show better properties than those of the nonwoven of Example 1. Compared with the nonwovens of Examples, the nonwoven of comparative Example 3, which does not satisfy the above requirements of the present invention, is poor in both bulkiness and strength.

10

### Claims

1. A continuous fiber nonwoven comprising composite continuous fibers having spiral crimps obtained by compositely spinning two thermoplastic resins having a difference in melting point of 15°C or more, characterised in that the contact points of the fibers are adhered to one another by fusing of the thermoplastic resin having the lower melting point and located on the outside of the spiral crimps.
2. A continuous fiber nonwoven according to claim 1, characterised in that the composite type of the composite continuous fibers is a parallel or eccentric sheath core type.
3. A method for producing a continuous fiber nonwoven comprising: preparing a first thermoplastic resin and a second thermoplastic resin having a melting point at least 15°C less than that of the first thermoplastic resin and an elastic shrinkage 1% less than that of the first thermoplastic resin; compositely spinning these resins in a composite ratio of 60/40 - 40/60 into a parallel type or an eccentric sheath core type, in which the second thermoplastic resin is a sheath and the first thermoplastic resin is a core eccentric to the sheath; stretching the resulting yarn over 1.2 times as long as the unstretched yarn at a temperature lower than the melting point of the second thermoplastic resin; and heat treating the yarn at a temperature higher than the melting point of the second thermoplastic resin and lower than the softening point of the first thermoplastic resin to adhere one to the other at the contact points of the fibers.
4. A method for producing a continuous fiber nonwoven according to claim 3, characterised in that the first thermoplastic resin is crystalline polypropylene and the second thermoplastic resin is high-density polyethylene.
5. A method for producing a continuous fiber nonwoven according to claim 3 or 4, characterised in that the heat treatment is conducted by a system of an oven with internal air circulation.
6. A method for producing a continuous fiber nonwoven according to claim 3 or 4, characterised in that the heat treatment is conducted by a hot pressing system.

40

45

50

55

(19)



Europäisches Patentamt  
European Patent Office  
Office européen des brevets



(11)

EP 0 747 521 A3

(12)

## EUROPEAN PATENT APPLICATION

(88) Date of publication A3:  
25.08.1999 Bulletin 1999/34

(51) Int Cl. 6: D04H 1/54, D04H 3/14

(43) Date of publication A2:  
11.12.1996 Bulletin 1996/50

(21) Application number: 96304104.1

(22) Date of filing: 05.06.1996

(84) Designated Contracting States:  
DE GB IT

(72) Inventor: Terakawa, Taiju  
Yasu-gun, Shiga-ken (JP)

(30) Priority: 06.06.1995 JP 16474995

(74) Representative: W.P. THOMPSON & CO.  
Eastcheap House

(71) Applicant: CHISSO CORPORATION  
Osaka-shi Osaka 530-6591 (JP)

Central Approach

Letchworth, Hertfordshire SG6 3DS (GB)

### (54) Continuous fiber nonwoven and method for producing the same

(57) A continuous fiber nonwoven comprising composite continuous fibers having spiral crimps obtained by compositely spinning two thermoplastic resins having a difference in the melting points of 15°C or more is

provided. It is characterised in that the contact points of the fibers are adhered to one another by fusing of the thermoplastic resin having the lower melting point and located on the outside of the spiral crimps.

EP 0 747 521 A3



European Patent  
Office

## EUROPEAN SEARCH REPORT

Application Number  
EP 96 30 4104

DOCUMENTS CONSIDERED TO BE RELEVANT			
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int.Cl.6)
A	US 4 269 888 A (EJIMA SHOZO ET AL) 26 May 1981 * column 3, line 6 - line 59; example 1 * ---	1-5	D04H1/54 D04H3/14
A	EP 0 391 260 A (CHISSO CORP) 10 October 1990 * column 4, line 26 - line 52; example 1 * -----	1-5	
TECHNICAL FIELDS SEARCHED (Int.Cl.6)			
D04H			
The present search report has been drawn up for all claims			
Place of search	Date of completion of the search	Examiner	
THE HAGUE	29 June 1999	V Beurden-Hopkins, S	
CATEGORY OF CITED DOCUMENTS			
X : particularly relevant if taken alone Y : particularly relevant if combined with another document of the same category A : technological background O : non-written disclosure P : intermediate document		T : theory or principle underlying the invention E : earlier patent document, but published on, or after the filing date D : document cited in the application L : document cited for other reasons & : member of the same patent family, corresponding document	

**ANNEX TO THE EUROPEAN SEARCH REPORT  
ON EUROPEAN PATENT APPLICATION NO.**

EP 96 30 4104

This annex lists the patent family members relating to the patent documents cited in the above-mentioned European search report.  
 The members are as contained in the European Patent Office EDP file on  
 The European Patent Office is in no way liable for these particulars which are merely given for the purpose of information.

29-06-1999

Patent document cited in search report		Publication date	Patent family member(s)	Publication date
US 4269888	A	26-05-1981	JP 927641 C JP 49075869 A JP 52012830 B DE 2358484 A GB 1446570 A US 4189338 A	13-10-1978 22-07-1974 09-04-1977 12-06-1974 18-08-1976 19-02-1980
EP 0391260	A	10-10-1990	JP 2269854 A JP 2849919 B DE 69010048 D DE 69010048 T US 5302220 A	05-11-1990 27-01-1999 28-07-1994 16-02-1995 12-04-1994